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(54) Title: HIGH SURFACE ENERGY POLYOLEFIN BLEND COMPOSITIONS (57) Abstract <p>Blended polymer compositions are described which comprise: (I) at least one crystalline alpha-olefin polymer; (II) from about 5 to about 60 % by weight of at least one elastomeric copolymer of an alpha-olefin and an alpha,beta-unsaturated carboxylic acid ester; (III) from about 1 to 30 % by weight of the product of the reaction of an alpha-olefin polymer and an alpha,beta-olefinically unsaturated carboxylic acid reagent; and (IV) from about 0 to about 40 % by weight of at least one modifier selected from the group consisting of (a) an ethylene-propylene copolymer elastomer which is different from the alpha-olefin polymer (I); (b) an elastomer which is a copolymer of a vinyl aromatic compound and a conjugated diene; and (c) a selectively hydrogenated block copolymer of a vinyl aromatic compound and a conjugated diene. The blended polymer compositions of the present invention exhibit improved surface properties such as adherability and paintability. In one embodiment, the blended polymer compositions can be painted directly without primer or adhesion promoter.</p>		

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HIGH SURFACE ENERGY POLYOLEFIN BLEND COMPOSITIONS

Field of the Invention

This invention relates generally to blended polymer compositions which comprise a mixture of at least one crystalline alpha-olefin polymer, an elastomeric copolymer of an alpha-olefin and an alpha,beta-unsaturated carboxylic ester, and the product of the reaction of an alpha-olefin polymer and an alpha,beta-olefinically unsaturated carboxylic acid reagent. The invention also relates to shaped articles prepared from the blended polymer compositions of the present invention which are receptive and adherent to protective and decorative coatings such as paints or printing inks.

Background of the Invention

Crystalline polyolefins such as polyethylene and polypropylene, and polymer blends containing such crystalline polyolefins have been widely used in many molding and extrusion applications. These polymers and polymer blends can be formed or shaped into lightweight and durable articles such as automobile parts, toys, housings for various types of equipment, etc. However, since these polymers and polymer blends are primarily non-polar, they generally are characterized as exhibiting poor adhesion to, for example, paint. Such polymers and polymer blends generally cannot be printed or painted without pretreatment, and they often exhibit poor stress crack-resistance.

The problem of paint adhesion has been a particular concern in the case of articles made from blends derived from thermoplastic olefin ("TPO") elastomeric compositions. In particular, TPO compositions which are blends of synthetic rubber and polyolefins such as polypropylene and polyethylene are used for fabricating

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lightweight and durable products for use in the automotive industry and in various other applications. It is important, therefore, that such articles can be painted so that there will be no significant visible difference between the painted TPO articles and the painted metallic parts of the vehicles.

Various procedures have been suggested for making articles containing substantial amounts of polypropylene and/or rubber more paint receptive. Adhesion promoters such as chlorinated polyolefins have been suggested in U.S. Patent 4,439,573.

It has also been suggested that when colored polymer blends are desired, the polyolefin blends can be melt-blended with an appropriate pigment. In some instances, pigmented materials have been found to be unsatisfactory for molding large parts and streaks form at the weld lines in the mold piece. Other undesirable effects are often observed.

Polymeric compositions are described in U.S. Patent 3,410,928 which are polyolefin blends comprising an ethylene or propylene homopolymer or an ethylene copolymer with an ethylene/acrylic acid copolymer. The blends are reported to exhibit improved adhesivity, printability, grease-resistance and stress crack-resistance when compared to the olefin polymers alone.

U.S. Patent 3,746,676 describes polyolefin compositions which comprise (1) about 35 to 92% of a crystalline polymer of an alpha-olefin having 2 to 8 carbon atoms containing at least 90% alpha-olefin units and at least part of which has been chemically combined with maleic anhydride, (2) 0 to 30% of a crystalline ethylene polymer, (3) about 5 to about 20% of an ethylene-propylene copolymer rubber containing 20 to 80% ethylene units, and (4) from about 3 to about 15% of rosin. These compositions are described as having excellent adhesion to paints, lacquers and inks without the need of expensive primers or other special surface treatments.

U.S. Patent 4,303,697 describes a process for modifying the surfaces of polyolefins to improve the adhesion of paint. The articles may be made of polyethylene, polypropylene, crystalline copolymers of ethylene with propylene, and

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blends thereof with elastomeric ethylene/propylene copolymers or with EPDM copolymers. The articles are prepared for painting by priming with a chlorinated polymeric material selected from chlorinated polypropylene and chlorinated polypropylene containing carboxylic anhydride groups, and thereafter exposing the primed article to ultraviolet radiation. The treatment improves both dry and wet adhesion of paint to the surfaces of the article.

U.S. Patent 4,888,391 describes TPO blends which are especially suited for use as flexible automotive body parts and other applications requiring reduced flexural modulus while maintaining good impact-resistance and tensile strength. The polymer blends described in this patent comprise a blend of polyolefin as the continuous phase with an ethylene/acrylate/acrylic acid terpolymer as the dispersed phase.

Thermoplastic compositions and articles made therefrom and which have a high receptivity to automotive paints are described in two PCT published applications identified as WO86/04912 and WO88/07564. WO86/04912 describes blends of (A) a copolymer of an ethylenically unsaturated carboxylic acid and ethylene, (B) an elastomer compatible with components (A) and (C), and (C) a crystalline polymer or copolymer of propylene. The WO88/07564 (corresponding to EP 357,643) describes a blend of (A) a copolymer of an ethylenically unsaturated carboxylic acid and ethylene, (B) an elastomer comprising an ethylene-alpha-olefin copolymer, (C) a crystalline homopolymer or copolymer of propylene, (D) an inorganic filler, and (E) a polyethylene or copolymer of ethylene and an alpha-olefin. Articles prepared from the thermoplastic compositions in both of these published patent applications are reported to exhibit a broad range of stiffness values and high impact and tensile strengths suitable for automotive applications such as bumpers, etc.

In European Patent Application 0312664, impact-resistant thermoplastic compositions having improved surface properties are described which comprise (A) a polypropylene resin, (B) a terpolymer of ethylene, alkyl acrylate or alkyl methacrylate, and an unsaturated dicarboxylic acid anhydride unit, and (C) an ethylene-propylene and/or ethylene-propylene non-conjugated diene terpolymer

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rubber. Among the improved surface properties described are improved adhesion, paintability and printability.

Summary of the Invention

Blended polymer compositions are described which comprise:

- (I) at least one crystalline alpha-olefin polymer;
- (II) from about 5 to about 60% by weight of at least one elastomeric copolymer of an alpha-olefin and an alpha,beta-unsaturated carboxylic acid ester;
- (III) from about 1 to 30% by weight of the product of the reaction of an alpha-olefin polymer and an alpha,beta-olefinically unsaturated carboxylic acid reagent; and
- (IV) from about 0 to about 40% by weight of at least one impact modifier selected from the group consisting of
 - (a) an ethylene-propylene copolymer elastomer which is different from the alpha-olefin polymer (I);
 - (b) an elastomer which is a copolymer of a vinyl aromatic compound and a conjugated diene; and
 - (c) a selectively hydrogenated block copolymer of a vinyl aromatic compound and a conjugated diene.

The blended polymer compositions of the present invention exhibit improved surface properties such as adhesivity and paintability. In one preferred embodiment, the blended polymer compositions are painted directly without primer or adhesion promoter.

Description of the Preferred Embodiments

(I) The Olefin Polymers.

One of the essential components of the blended polymer compositions of the present invention is at least one olefin polymer, and in one embodiment, the olefin polymer is present in the blended polymer composition in amounts of at least about 20% by weight and up to about 80% by weight. In another embodiment, the polymer compositions of the present invention will contain from about 25% to about 50% by weight of the olefin polymer.

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The olefin polymers employed in the blends of the present invention generally are semi-crystalline or crystallizable olefin polymers including homopolymers, copolymers, terpolymers, or mixtures thereof, etc., containing one or more monomeric units. Polymers of alpha-olefins or 1-olefins are preferred in the present invention, and these alpha-olefins may contain from 2 to about 20 carbon atoms. Alpha-olefins containing 2 to about 6 carbon atoms are preferred. Thus, the olefin polymers may be derived from olefins such as ethylene, propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 1-octene, 1-decene, 4-ethyl-1-hexene, etc., or mixtures of two or more of these olefins. Examples of particularly useful olefin polymers include low-density polyethylene, high-density polyethylene, linear low density polyethylene, ultra low density polyethylene, polypropylene (including isotactic polypropylene), (high and low density) poly(1-butene), poly(4-methyl-1-pentene), ultra low molecular weight polyethylene, ethylene-based ionomers, poly(4-methyl-1-pentene), ethylene-propylene copolymers, ethylene-propylene-diene copolymers (EPDM), copolymers of ethylene and/or propylene with other copolymerizable monomers such as a copolymer of ethylene-1-butylene. Halogenated olefins, polymers and copolymers may also be used in this invention.

The number average molecular weight of the polyolefins is preferably above about 10,000 and more preferably above about 50,000. In addition, it is preferred in one embodiment that the apparent crystalline melting point be above about 75°C and preferably between about 75°C and about 250°C. Most commercial polyethylenes have a number average molecular weight of from about 50,000 to about 500,000. The olefin polymers useful in preparing the polymer blends of the present invention are well-known to those skilled in the art and many are available commercially. The olefin polymers may be homopolymers, impact copolymers, block copolymers, random copolymers, thermoplastic olefinic elastomers (TPO), etc., or mixtures thereof. Polyethylene and polypropylene are preferred olefin polymers. High density polyethylenes such as Phillips 6001, Cain's 7040 and Soltex G5012, and polypropylene homo- and copolymers such as Himont's Profax 6523 and 8623,

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Shell's 7C54H and 7C06 or Exxon's PP1012, PD7132 and PD7163, or Aristech's 4040F are particularly preferred.

II. Elastomer Copolymer of Alpha-Olefin and Alpha,Beta-Unsaturated Carboxylic Acid Ester.

The blended polymer compositions of the present invention contain from about 5 to about 60% by weight of at least one elastomeric copolymer of an alpha-olefin and an alpha,beta-unsaturated carboxylic acid ester. The alpha-olefins used in the preparation of the elastomer (II) may be any of the alpha-olefins described as being useful and preparing the olefin polymer (I) described above. In one preferred embodiment, the alpha-olefins contain from 2 to about 6 carbon atoms and more preferably, the alpha-olefin is ethylene, propylene or a mixture of ethylene and propylene.

The alpha,beta-unsaturated carboxylic acid esters useful in the preparation of the elastomer (II) may be derived from monocarboxylic as well as dicarboxylic acids although the monocarboxylic acids are preferred. The monocarboxylic acid esters may be exemplified by the following formula



wherein R is hydrogen or an unsaturated aliphatic, alicyclic, aryl, alkaryl or heterocyclic group; R' is hydrogen or an alkyl group containing from 1 to about 4 carbon atoms such as a methyl or ethyl group; and R'' is an alkyl group containing from 1 to about 10 carbon atoms and which may also contain functional groups such as hydroxyl, epoxy, double bonds, etc. Preferably, R is hydrogen or an alkyl group containing from 1 to about 10 carbon atoms, and more preferably, R is hydrogen. Specific examples of esters characterized by the above formula which are useful in forming the elastomers (II) include methyl acrylate, ethyl acrylate, butyl acrylate, methyl methacrylate, ethyl methacrylate, ethyl ethacrylate, etc. A preferred ester is methyl acrylate.

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Copolymers of α -olefins and other carboxylic acid esters such as those characterized by the formula



where R is an alkyl group and can be used as elastomer copolymer (II). For example the copolymer may be derived from ethylene and vinyl acetate (EVA).

The elastomeric copolymer (II) may comprise from about 40 to about 95% by weight of the alpha-olefin and from about 5 to about 60% by weight of the carboxylic acid ester. In another embodiment the copolymers (II) contain from about 15 to about 25% by weight of the carboxylic ester and about 75 to about 85% of ethylene. The copolymers can be prepared by procedures well known to those skilled in the polymer art, and such elastomers are available commercially. Examples of specific elastomers (II) include ethylene methyl acrylate copolymers (EMAC) such as those commercially available from Chevron under the designations SP2205, SP2255 and SP2260 having methyl acrylate contents of 20%, 18% and 24% by weight, respectively; from Exxon under the general trade designation "Escor" and available in commercial grades designated, e.g., TC110 and TC112 containing 20% and 18% methyl acrylate, respectively. Developmental Escor grade copolymers are also available from Exxon containing from 6% by weight of methyl acrylate (XS 11.04) to 28% by weight of methyl acrylate (XS 21.04 and 55.48). Lotryl 3610 is an ethylene-methyl acrylate copolymer having a methyl acrylate content of 29% by weight and is available from Atochem, Inc. Some developmental EMAC are also available from Chevron under trade designations such as TD1956, TD1967, TD1972 and another TD1972, and these contain 6%, 35%, 42% and 60% by weight of methyl acrylate, respectively.

Also useful as elastomer (II) are ethylene ethyl acrylates (EEA) such as those available from Union Carbide under the Bakelite designations DPD-6182, DPD-6169 and DPDA 9169. EBAs (ethylene butyl acrylates) are available from Atochem Inc. under designations such as 17BG04 (15-18% ester content) and 30BA02 (28-32% ester content).

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III. Reaction Product of Alpha-Olefin Polymer and Alpha-Beta Olefinically Unsaturated Carboxylic Acid Reagent.

The blended polymer compositions of the present invention also contain the product of the reaction of at least one alpha-olefin polymer and an alpha,-beta-olefinically unsaturated carboxylic acid reagent. The reaction product (III) will be present in an amount of from about 1 to about 30% by weight based on the total weight of polymer composition. Preferably, the alpha-olefin polymer is derived from an aliphatic alpha-olefin such as ethylene, propylene, 1-butene, etc. The olefin polymers are preferably polyethylenes, polypropylenes or mixtures thereof. The number average molecular weight of the polyolefins utilized in the reaction with the carboxylic acid reagent is generally lower than the number average molecular weight of the polyolefins utilized as a major component in the blended polymer compositions of the present invention and described above as component (I). In one embodiment, the number average molecular weights of the polyolefins utilized in this component may range from about 1000 to about 10,000 or higher.

The amount of carboxylic acid reagent reacted with the polyolefin may range from about 0.1% to about 30% by weight based on the weight of the olefin polymer. More generally, the amount of acid reagent reacted with the polyolefin will be less than 10% and most often between 0.1 and 5% by weight. The alpha,-beta-olefinically unsaturated carboxylic acid reagents utilized to form the reaction product (III) may be mono- or dicarboxylic acid reagents.

The monobasic alpha,beta-olefinically unsaturated carboxylic acid reagents are carboxylic acids or derivatives corresponding to the formula



wherein R is hydrogen or a saturated aliphatic or alicyclic, aryl, alkaryl or heterocyclic group. Preferably, R is hydrogen or an alkyl group containing from 1 to about 10 carbon atoms. R₁ is hydrogen or an alkyl group containing from 1 to about 10 carbon atoms. The total number of carbon atoms in R and R₁ should not

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exceed 18 carbon atoms. Specific examples of useful monobasic carboxylic acid derivatives include acrylic acid, methacrylic acid, cinnamic acid, crotonic acid, sodium acrylate, calcium acrylate and magnesium acrylate, etc.

Examples of dicarboxylic acids and useful derivatives thereof include maleic acid, maleic anhydride, fumaric acid, mesaconic acid, himic anhydride, itaconic acid, citraconic acid, itaconic anhydride, citraconic anhydride, monomethyl maleate, monosodium maleate, etc. A particularly preferred alpha,beta-olefinically unsaturated carboxylic reagent is maleic acid anhydride. The maleic acid anhydride may be used with other copolymerizable monomers such as maleimide and bis-maleimide, or with other monomers to introduce functional groups such as alcohols, mercaptans, silanols, epoxy compounds, oxazoline and the salts thereof.

The reaction between the carboxylic acid reagent in the polyolefin can be effected by means known to those skilled in the art. For example, the reaction can be conducted in solutions by a melt process in the presence of a free radical initiator. The free radical initiators usually are either peroxides or various organic azo compounds. The amount of initiator utilized generally from about 0.01% to about 5% by weight based on the combined weight of the olefin and the carboxylic reagent.

An example of a commercially available maleic acid anhydride grafted polypropylene is Epolene E-43 wax from Eastman Chemical Products, Inc. Epolene E-43 has an acid number of 47. Epolene C-16 and C-18 waxes are maleic acid grafted polyethylenes with approximate molecular weights of 8000 and 4000, respectively. Maleated ethylene-propylene elastomers also are useful, and such elastomers are available from Exxon Chemical Company under identification numbers 99-10, 99-14 and 99-26. These copolymers contain 77%, 64% and 43% of ethylene, respectively, and the maleated products contain 0.76%, 0.56% and 0.35% of maleic acid or anhydride, respectively.

Other examples of commercially available maleic acid anhydride grafted polyolefins include Kraton FG1901X from Shell which is often referred to as a maleated selectively hydrogenated SEBS copolymer; terpolymers available from

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CdF Chimie under designations such as Lotader 3200 (prepared from a mixture of 88% by weight ethylene, 9% by weight butyl acrylate and 3% maleic anhydride), Lotader 6600 (70% ethylene, 27% acrylic ester and 3% maleic anhydride) etc.; ethylene vinyl acetate copolymers grafted with maleic anhydride (EVA-MAH) are available from Quantum Chemical Corp.

IV. Modifiers.

The blended polymer compositions of the present invention may also contain at least one modifier which improves various properties such as low temperature and room temperature impact strength. The amount of modifier incorporated into the polymer blends of the present invention may vary from about 0.1% to about 30% by weight based on the total weight of the polymer composition. In another embodiment, the polymer blends of the present invention contain from about 1 to about 20% and more preferably from about 5 to about 15% by weight of the modifier.

IVa. Ethylene-Propylene Copolymer Elastomer.

The polymer blends of the present invention may contain, as modifiers, elastomeric copolymers of ethylene and propylene which are different from the alpha-olefin polymers (I). These elastomers may generally comprise from about 20 to about 80 weight percent of ethylene units. In addition to the elastomers containing only ethylene and propylene (generally referred to as ethylene-propylene rubber or EPR), the ethylene-propylene copolymers may also contain a diene as a third monomer. Generally, the dienes are non-conjugated dienes such as methyl tetrahydroindene, 5-vinyl norbornene, 5-ethylidene-2-norbornene, dicyclopentadiene, 1,4-hexadiene, 1,6-octadiene, etc. These types of elastomers are referred to in the art as EPDM.

The elastomers may be partially crosslinked with numerous crosslinking agents such as organic peroxides (e.g., dicumyl peroxide), divinyl benzene or bis-maleimide or multi-functional maleimide (functionality >2).

Ethylene-propylene elastomers and EPDM elastomers are available commercially. The Exxon Chemical Company has a variety of commercially available ethylene/propylene copolymer elastomers available under the general trade

designation "Vistalon". For example, Vistalon ECA 8232 is a commercial ethylene/propylene copolymer elastomer from Exxon having an ethylene content of about 67% by weight and a Mooney viscosity of about 32 at 125°C. Vistalon 719 is a commercially available pellet form ethylene/propylene rubber from Exxon. Examples of an EPDM rubbers available from Exxon include Vistalon 1721 and Vistalon 3708. An ethylene/propylene rubber also is available from Polysar under the designation Polysar 306, and a useful EPDM also is available from DuPont under the general trade designation Nordel 2722.

Masterbatches containing impact modifying elastomers also are useful. Such masterbatches also provide for improved processing, dispersion and ease of handling (e.g., pellets). Polyolefin/EPR masterblends from Enimont under the trade designation Dutral are examples of useful masterbatches. For example, PM 06PLE is a masterblend of Dutral CO-059 in HDPE. CO-059 is an EPR containing ethylene/propylene in a weight ratio of 57/43.

IVb. Copolymer of a Vinyl Aromatic Compound and a Conjugated Diene.

The blended polymer composition of the present invention also may contain as modifier, an elastomer which is a copolymer of a vinyl aromatic compound and a conjugated diene. Examples of vinyl aromatic hydrocarbons which may be utilized to prepare the elastomers include styrene and the various substituted styrenes such as o-methyl styrene, p-methyl styrene, p-tert-butyl styrene, 1,3-dimethyl styrene, alpha-methyl styrene, beta-methyl styrene, p-isopropyl styrene, 2,3-dimethyl styrene, o-chloro styrene, p-chloro styrene, o-bromo styrene, 2-chloro-4-methyl styrene, etc. A preferred vinyl aromatic hydrocarbon is styrene.

The conjugated dienes which may be utilized to prepare the elastomers of (IVb) are those containing from 4 to about 10 carbon atoms and more generally, from 4 to 6 carbon atoms. Examples include 1,3-butadiene, 2-methyl-1,3-butadiene (isoprene), 2,3-dimethyl-1,3-butadiene, 2-chloro-1,3-butadiene, 1,3-pentadiene, 1,3-hexadiene, etc. Mixtures of these conjugated dienes also may be used. The preferred conjugated dienes are isoprene and 1,3-butadiene.

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The elastomers (IVb) may be random copolymers or block copolymers. Block copolymers of vinyl aromatic hydrocarbons and conjugated dienes which can be used as modifier (IVb) are described more fully below when describing the modifier (IVc).

Useful copolymer elastomers of vinyl aromatic compounds and conjugated dienes are available commercially. For example, several styrene butadiene rubbers are available commercially from the Firestone Tire and Rubber Company such as the elastomers identified by the trade designations Stereon 840A and Stereon 721. Fina Oil and Chemical Co. offers styrene butadiene rubbers as Finaprene 401, 414, 417 and 480, for example. Finaprene 414 contains about 40% styrene and Finaprene 417 contains 30% styrene.

IVc. Selectively Hydrogenated Block Copolymers of a Vinyl Aromatic Compound and a Conjugated Diene.

The modifiers which may be utilized herein include the selectively hydrogenated block copolymers of a vinyl aromatic hydrocarbon and a conjugated diene.

The block copolymers of vinyl aromatic hydrocarbons and conjugated dienes which may be utilized in the polymer blends of the present invention include any of those which exhibit elastomeric properties and those which have 1,2-micro-structure contents prior to hydrogenation of from about 7% to about 100%. The block copolymers may be diblock, triblock, multiblock, starblock, polyblock or graftblock copolymers. Throughout this specification and claims, the terms diblock, triblock, multiblock, polyblock, and graft or grafted-block with respect to the structural features of block copolymers are to be given their normal meaning as defined in the literature such as in the Encyclopedia of Polymer Science and Engineering, Vol. 2, (1985) John Wiley & Sons, Inc., New York, pp. 325-326, and by J.E. McGrath in Block Copolymers, Science Technology, Dale J. Meier, Ed., Harwood Academic Publishers, 1979, at pages 1-5.

Such block copolymers may contain various ratios of conjugated dienes to vinyl aromatic hydrocarbons including those containing up to about 60% by weight

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of vinyl aromatic hydrocarbon. Accordingly, multi-block copolymers may be utilized which are linear or radial symmetric or asymmetric and which have structures represented by the formulae A-B, A-B-A, A-B-A-B, B-A-B, $(AB)_{0,1,2...}BA$, etc., wherein A is a polymer block of a vinyl aromatic hydrocarbon or a conjugated diene/vinyl aromatic hydrocarbon tapered copolymer block, and B is a polymer block of a conjugated diene.

The block copolymers may be prepared by any of the well-known block polymerization or copolymerization procedures including sequential addition of monomer, incremental addition of monomer, or coupling techniques as illustrated in, for example, U.S. Patents 3,251,905; 3,390,207; 3,598,887; and 4,219,627. As well known, tapered copolymer blocks can be incorporated in the multi-block copolymers by copolymerizing a mixture of conjugated diene and vinyl aromatic hydrocarbon monomers utilizing the difference in their copolymerization reactivity rates. Various patents describe the preparation of multi-block copolymers containing tapered copolymer blocks including U.S. Patents 3,251,905; 3,639,521; and 4,208,356, the disclosures of which are hereby incorporated by reference.

Conjugated dienes which may be utilized to prepare the polymers and copolymers are those containing from 4 to about 10 carbon atoms and more generally, from 4 to 6 carbon atoms. Examples include from 1,3-butadiene, 2-methyl-1,3-butadiene (isoprene), 2,3-dimethyl-1,3-butadiene, chloroprene, 1,3-pentadiene, 1,3-hexadiene, etc. Mixtures of these conjugated dienes also may be used. The preferred conjugated dienes are isoprene and 1,3-butadiene.

Examples of vinyl aromatic hydrocarbons which may be utilized to prepare the copolymers include styrene and the various substituted styrenes such as o-methylstyrene, p-methylstyrene, p-tert-butylstyrene, 1,3-dimethylstyrene, alpha-methylstyrene, beta-methylstyrene, p-isopropylstyrene, 2,3-dimethylstyrene, o-chlorostyrene, p-chlorostyrene, o-bromostyrene, 2-chloro-4-methylstyrene, etc. A preferred vinyl aromatic hydrocarbon is styrene.

Many of the above-described block copolymers of conjugated dienes and vinyl aromatic compounds are commercially available. The number average

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molecular weight of the block copolymers, prior to hydrogenation, is from about 20,000 to about 500,000, preferably from about 40,000 to about 300,000.

The average molecular weights of the individual blocks within the copolymers may vary within certain limits. In most instances, the vinyl aromatic block will have a number average molecular weight in the order of about 2000 to about 125,000, and preferably between about 4000 and 60,000. The conjugated diene blocks either before or after hydrogenation will have number average molecular weights in the order of about 10,000 to about 450,000 and more preferably from about 35,000 to 150,000.

Also, prior to hydrogenation, the vinyl content of the conjugated diene portion is from about 10 to about 80%, and the vinyl content is preferably from about 25 to about 65%, particularly 35 to 55% when it is desired that the modified block copolymer exhibit rubbery elasticity. The vinyl content of the block copolymer can be measured by means of nuclear magnetic resonance.

Specific examples of diblock copolymers include styrene-butadiene, styrene-isoprene, and the hydrogenated derivatives thereof. Examples of triblock polymers include styrene-butadiene-styrene, styrene-isoprene-styrene, alpha-methylstyrene-butadiene-alpha-methylstyrene, and alpha-methylstyrene-isoprene-alpha-methylstyrene.

The selective hydrogenation of the block copolymers may be carried out by a variety of well known processes including hydrogenation in the presence of catalysts such as Raney nickel, noble metals such as platinum, palladium, on support systems such as charcoal, etc., and soluble transition metal catalysts. Suitable hydrogenation processes which can be used are those wherein the diene-containing polymer or copolymer is dissolved in an inert hydrocarbon diluent such as cyclohexane and hydrogenated by reacting with hydrogen in the presence of a soluble hydrogenation catalyst. Such procedures are described in U.S. Patents 3,113,986 and 4,226,952, the disclosures of which are incorporated herein by reference. Such hydrogenation of the block copolymers which are carried out in a manner and to extent as to produce selectively hydrogenated copolymers having a residual

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unsaturation content in the polydiene block of from about 0.5 to about 20% of their original unsaturation content prior to hydrogenation.

In one embodiment, the conjugated diene portion of the block copolymer is at least 90% saturated and more often at least 95% saturated while the vinyl aromatic portion is not significantly hydrogenated. Particularly useful hydrogenated block copolymers are the hydrogenated block copolymers of styrene-isoprene-styrene such as an (ethylene/propylene)-styrene block polymer. When a styrene-butadiene-styrene block copolymer is hydrogenated, it is desirable that the 1,2-polybutadiene to 1,4-polybutadiene ratio in the polymer is from about 30:70 to about 70:30. When such a block copolymer is hydrogenated, the resulting product resembles a regular copolymer block of ethylene and 1-butene (EB). As noted above, when the conjugated diene employed as isoprene, the resulting hydrogenated product resembles a regular copolymer block of ethylene and propylene (EP). One example of a commercially available selectively hydrogenated copolymer is Kraton G-1652 which is a hydrogenated SBS triblock comprising 30% styrene end blocks and a mid-block equivalent is a copolymer of ethylene and 1-butene (EB). This hydrogenated block copolymer is often referred to as SEBS.

In another embodiment, the selectively hydrogenated block copolymer is of the formula



wherein $n = 0$ or 1 ;
 o is 1 to 100 ;
 p is 0 or 1 ;

each B prior to hydrogenation is predominantly a polymerized conjugated diene hydrocarbon block having a number average molecular weight of about 20,000 to about 450,000;

each A is predominantly a polymerized vinyl aromatic hydrocarbon block having a number average molecular weight of from about 2000 to about

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115,000; the blocks of A constituting about 5% to about 95% by weight of the copolymer; and the unsaturation of the block B is less than about 10% of the original unsaturation. In other embodiments, the unsaturation of block B is reduced upon hydrogenation to less than 5% of its original value, and the average unsaturation of the hydrogenated block copolymer is reduced to less than 20% of its original value. Maleated Kraton, for example, Kraton FG1901 is obtained by maleating the hydrogenated Kraton rubbers.

V. Pigment.

The blended polymer composition of the present invention may also contain at least one pigment when it is desired to impart color to the polymer composition. The amount of pigment included in the compositions will be an amount sufficient to provide the desired color to the polymer composition, and amounts of up to about 20% by weight or more, based on the total weight of polymer composition, can be utilized. The pigments may be incorporated directly into the blended polymer composition in dry form, color concentrates or as a predispersion containing one or more pigment.

Liquid colorant systems are also useful for coloring the blended polymer compositions of this invention. For example, Spectraflo™ custom colorant systems available from Ferro Corporation can be used.

The pigments which may be incorporated into the polymer compositions of the present invention may be organic pigments and/or inorganic pigments. The term "pigment" as used in the specification and claims also includes some materials which are sometimes referred to as fillers and polymers provided that these materials have an effect on the color, and these include metal powders, metal oxides and other inorganic compounds. Metal powders such as aluminum or bronze may be utilized as pigments to produce desired colors as well as provide other properties including improved thermal properties and electrical conductivity. Among the metallic oxides which can be utilized as pigments are zinc oxide, aluminum oxide, magnesium oxide, iron oxide red, iron oxide yellow, chrome oxide green, and titanium dioxide. Other inorganic pigments which may be utilized to provide color

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to polymeric materials include zinc sulfide, cadmium sulfoselenide, cadmium mercury, zinc chromate, cobalt aluminate, chrome cobalt alumina, ultra-marine blue and lead carbonate.

Carbon blacks are well known color pigments often utilized in polymer formulations. Among the carbon blacks which may be utilized in the present invention are furnace blacks, channel blacks and lamp blacks. More specifically, examples of carbon blacks include super abrasion furnace (SAF) blacks, high abrasion furnace (HAF) blacks, fast extrusion furnace (FEF) blacks, fine furnace (FF) blacks, intermediate super abrasion furnace (ISAF) blacks, medium procession channel blacks and conductive blacks. Other carbon blacks which may be utilized include acetylene blacks. Mixtures of two or more of the above blacks may also be used. Carbon black concentrates are particularly useful. The concentrates generally comprise a high concentration of one or more carbon blacks in a carrier such as a resin. For example, Colonial 3430 is a black color concentrate of 30% N-110 carbon black, 1% of an antioxidant and 69% of polypropylene as a carrier, and Colonial 4400 is a concentrate of 40% carbon black and 60% of linear low density polyethylene. These concentrates are available from Colonial Rubber Works Inc., Dyersburg, TN. Other resin carriers include, EVA, HDPE, LDPE and polyethylene.

A wide variety of organic pigments can be utilized as colorants in the polymeric compositions of the present invention. Some typical organic pigment colorants include Para Red, Lithol Rubine, Helio Bordeaux, Thio Indigo, Thio Indigoid, Toluidine, Dioxazine, Red Lake C, Red Lake R, etc.

V. Fillers and Fibers.

The blended polymer compositions of the present invention may contain one or more fillers of the type used in the polymer art. Examples of fillers employed in a typical compounded polymer blend according to the present invention included talc, calcium carbonate, mica, wollastonite, dolomite, glass fibers, cellulose fibers, PET fibers, boron fibers, carbon fibers, glass beads, or mixtures thereof. A naturally occurring black solid crude hydrocarbon asphaltene filler is available from Geoplas, Inc., Granville, Ohio, under the designation Geoplas™ GP Powder. Preferred fillers

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include commercially available talc such as R.T. Vanderbilt's Select-A-Sorb and glass fibers. The amount of filler and fibers included in the blended polymer compositions of the present invention may vary from about 1% to about 70% of the combined weight of polymer in filler. Generally, amounts of from 5% to 30% are utilized.

The fillers and fibers may be treated with coupling agents to improve the bond between the fillers and fibers to the resin. For example, the fillers can be treated with materials such as fatty acids (e.g., stearic acid), silanes, maleated polypropylene, etc. The amount of coupling agents used is an amount effective to improve the bond between the fillers and fibers with the resin.

Other additives may be included in the polymer blends of the present invention to modify or to obtain desirable properties. For example, stabilizers, lubricants, nucleating agents, plasticizers, mold-release agents, and inhibitors of oxidative, thermal and ultraviolet light degradation may be included in the polymer blends may be included in the polymer blends.

The stabilizers can be incorporated into the composition at any stage in the preparation of the polymer blends, and preferably, the stabilizers are included early to preclude the initiation of degradation before the composition can be protected. The oxidative and thermal stabilizers useful in the polymer blends of the present invention include those used in polymers generally. They include, for example, up to about 1% by weight, based on the weight of the polymer blend, of Group I metal halides such as sodium, potassium, lithium and cuprous halides (e.g., chloride, bromide, and iodide), hindered phenols, hydroquinones, and various substituted derivatives of these materials and combinations thereof. Specific examples of hindered phenolic stabilizers include a family of hindered phenols available commercially from Ciba-Geigy under the trade designation "Irganox" such as Irganox 1010 and Irganox 1076. Irganox 1010 is reported to be tetrakis [methylene (3,5-di-tert-butyl-4-hydroxy) hydrocinnamate] methane. Irganox 1076 is n-octadecyl 3-(3,5-di-tert-butyl-4-hydroxy-phenyl) propionate.

The ultraviolet light stabilizers may be included in amounts of up to about 2% by weight based on the weight of the polymer blend. Examples of ultravio-

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let light stabilizers include various substituted resorcinols, salicylates, benzotriazoles, benzophenones, etc. For certain applications, molded parts made from this material may be partially or completely exposed to U.V. light.

Suitable lubricants and mold release agents may be included in amounts of up to about 1% by weight based on the weight of the polymer blend include materials such as stearic acid, stearic alcohol, stearamides. The plasticizers which may be included in amounts of up to about 20% by weight based on the weight of the polymer blend include materials such as dioctylphthalate, bibenzylphthalate, butylbenzophthalate, hydrocarbon oils, epoxidized tall oil, sulfonamides such as paratoluene ethyl sulfonamides, etc.

The blended polymer compositions of the present invention including components (I), (II), (III), and optionally (IV) and/or (V), and other conventional additives can be prepared by techniques well known to those skilled in the art. For example, a particularly useful procedure is to intimately mix the polymers using conventional mixing equipment such as a mill, a Banbury, a Brabender, a single or twin screw extruder, continuous mixers, kneaders, etc. For example, the polymers may be intimately mixed in the form of granules and/or powder in a high shear mixer. One preferred process for preparing the blended polymers utilizes the Farrel Continuous Mixer (FCM CP-23). Short residence times and high shear are readily obtained in a CP-23. "Intimate" mixing means that the mixture is prepared with sufficient mechanical shear and thermal energy to produce a dispersed phase which is finely divided and homogeneously dispersed in the continuous or principal phase.

The blended polymer compositions of the present invention may be prepared by melt-blending the olefin polymer (I) with from about 5% to about 60% by weight of the elastomeric copolymer (II), from about 1 to about 30% by weight of the reaction product (III) and from 0 to about 40% of the above-described modifiers IV. The polymer blends of the present invention are characterized as having improved adherability and paintability.

The blended polymer compositions of the invention are characterized by improved surface characteristics (high surface energy) such as enhanced

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paintability, printability, platability, adhesion to foam (e.g., polyurethane foam) metal (e.g., aluminum), plastic substrates, etc. For example, once the polymer composition has been formed into shaped articles, they can be painted with conventional paints or powder coatings, and in many instances, without any special pretreatment of the surface of the article such as the application of an adhesion promoters. Adhesion promoters are not required, and in some instances, adhesion promoters have a negative effect on the adhesion of paints to the surface. Epoxy, polyester or polyurethane powder coatings can be used to color the blended polymer compositions of the invention. For example, Vedoc® powder coatings and Impress™ premold powder coatings which are available from Ferro Corporation are useful for this purpose.

The blended polymer compositions of the present invention can be processed into shaped articles by extrusion, coextrusion, thermoforming, blow-molding, injection-molding, compression-molding, calendaring, laminating, stamping, pultrusion, etc. In particular, shaped articles can be prepared by thermoforming sheets of the blended polymer compositions of the present invention. The blended polymer compositions of the invention are recyclable.

When the blended polymer compositions contain pigments such as carbon black or the liquid color concentrates described above, it is possible to apply only a clear coat to the formed article and to obtain a satisfactory product. In other instances, typical and multilayer coatings can be applied to the formed articles prepared from the blended polymer compositions of the invention, and good adhesion of the coatings is observed. Thus, primer coatings, base coatings, and clear coatings can be applied with success by techniques normally used in the coating or paint industry such as spraying, brushing, dipping, etc.

Once applied, the coatings can be dried by heating to an elevated temperature of about 80°F to about 400°F. The dry film thickness of primer coatings deposited on substrates usually ranges from about 0.2 mil to about 1.5 mils although greater film thicknesses can be deposited but generally are unnecessary.

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After the primer coating composition has been applied to the substrate, a second coating composition, generally pigmented, can be applied to the primer-coated substrate which, for convenience, will be referred to herein either as a "top coat" when the second coating is the final coating, or as a "base coat" when a third coating composition (generally a clear coating composition is described hereinafter) is applied thereover. The pigmented coating composition can be applied over the primer coating wet-on-wet, that is, the primer-coated substrate can be subsequently coated without first being dried. Alternatively, the primer can be partially cured by baking prior to the application of the pigmented base coat. In one embodiment, for example, the primer coated substrate is baked for a period of from about 20 seconds to about 180 seconds at a temperature of about 80°F to about 400°F or higher prior to application of the pigmented base coat composition.

The pigmented base coat composition which is applied over the primer composition generally comprises at least one thermosetting resin, at least one pigment and at least one solvent. Preferably, the thermosetting resin is a cross-linkable thermosetting resin capable of forming chemical bonds with the thermoset resin contained in the primer coating. Examples of cross-linkable resins which can be utilized in the base coat include amino resins, alkyd resins, urethane resins, acrylic resins, epoxy resins, acrylic resins, phenolic resins, saturated or unsaturated polyester resins, allyl resins and furan resins. The choice of cross-linkable thermosetting resin utilized in the base coat composition can be readily determined by one skilled in the art based upon the properties which are desired. For example, if a soft-flexible film is desired, the polyester resins can be utilized, and if a harder film is desired, amino resins, acrylic resins or epoxy resins may be selected.

Any of the amino resins, alkyd resins, urethane resins, acrylic resins, epoxy resins, phenolic resins described earlier as resins useful in the primer coating compositions can be utilized in the base coat composition.

The polyester resin which can be utilized in the base coat compositions may be either saturated or unsaturated polyester resins formed by condensing the polycarboxylic acid or anhydride (either saturated or unsaturated) with at least one

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polyhydric alcohol. Examples of saturated polyester resins include the products of the reaction of a saturated dicarboxylic acid or anhydride such as phthalic acid or anhydride, isophthalic acid, tetrahydrophthalic anhydride, hexahydrophthalic anhydride, succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid and sebacic acid with a dihydric alcohol such as ethylene glycol, propylene glycol, butylene glycol, diethylene glycol, triethylene glycol, and neopentyl glycol. Small amounts of a polyhydric alcohol such as glycerol, pentaerythritol, trimethylolpropane or sorbitol may be used in combination with the glycol. The unsaturated polyester resins are characterized by the vinyl unsaturation and the polyester backbone. The unsaturated polyester resins are obtained by reaction of any of the above-described glycols and polyols with an unsaturated dicarboxylic acid or anhydride such as maleic anhydride, fumaric anhydride, chloromaleic acid, itaconic acid, citraconic acid and mesaconic acid.

The allyl resins are formed by the addition polymerization of compounds containing the allyl group ($\text{CH}_2=\text{CHCH}_2-$), such as esters of allyl alcohol and dibasic acids. Examples of allyl resins include diallylphthalate, diallylisophthalate, diallylmaleate and diallylchloredate.

The furan resins which may be utilized in the basecoat compositions are obtained primarily by the condensation polymerization of furfuryl alcohol in the presence of strong acids, sometimes in combination with formaldehyde or furfuraldehyde. These furan resins also include resins made by condensing phenol with furfural alcohol or furfuryl, and furfuryl-ketone polymers.

As mentioned above, the cross-linking thermosetting resin is incorporated into the basecoat compositions used in the present invention to improve the adhesion of the basecoat applied to the primer coating. The cross-linking resin forms bonds with any cross-linkable resins contained in the primer.

Pigments may be included in the basecoat formulation and any of the pigments described above (V) can be used in the basecoat. The choice of a particular pigment or pigments will be determined by one skilled in the art from a consideration of the characteristics and color desired for the basecoat. The solvents may be

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aliphatic and/or aromatic solvents of the types normally used in paints. Also, as in the primer coating, conventional additives may be included in the basecoat formulation such as surfactants, antioxidants, ultraviolet light absorbers, stabilizers, rheology control agents, coalescing agents, levelling agents, etc.

In another embodiment, a clear coating composition is applied to the substrate as formed, to the substrate containing the primer coat, or to the substrate which has been given a primer coating and a basecoat. The clear coating composition can also be applied wet-on-wet over the primer or basecoat or, alternatively, the basecoat composition can be partially cured by baking prior to the application of the clear coat. In one preferred embodiment, the basecoat is baked for a period of about 20 seconds to about 180 seconds at a temperature of from 80°F to about 400°F or higher prior to application of the clear topcoat composition.

The clear coating composition comprises at least one solvent and at least one thermosetting resin, and the clear coating compositions can be formulated with the same types of solvents and additives as have been described above in connection with the primer coating composition. However, the clear compositions do not contain pigments. After the clear coating composition is applied to the substrate, the coated substrate is heated to a temperature sufficient to cure the coated substrate. Generally, temperatures of from about 80°F to about 400°F are sufficient to effect the cure. This final heating step removes the solvent present and causes the coating layers to fuse together in a bonding process. The coated substrates prepared in this manner and in accordance with the process and compositions of the present invention are characterized by superior adhesion of the coatings, abrasion resistance and resistance to weathering and industrial pollutants.

As with the primer coatings, the basecoat and clear coat compositions can be applied to the substrate by techniques well known in the art. For example, the coating compositions can be applied by spraying, dipping, brushing, etc. The dry film thickness of the basecoat may range from about 0.5 mil to about 2 mils, and the dry film thickness of the clear topcoat may range from about 0.2 mil to about 2 mils.

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The improved adhesion to paint exhibited by the blended polymer composition of the present invention is demonstrated in some of the following examples with injection molded test plaques of the polymer compositions which are cleaned with a soft cloth dampened with methyl ethyl ketone and air dried at room temperature. Immediately after cleaning, the plaque is primed by spray coating with two coats of urethane primer HSE 9440 (from PPG Industries) to provide a dry coating thickness of 1.0 to 2.0 mil. There is a two-minute flash drying period between coats and a ten-minute flash drying period after the second coat to evaporate low boiling solvents in a dust-free chamber at room temperature. The coatings on the plaques are then cured by baking at 250°F (121°C) for 30 minutes.

The primed plaque is then provided with a basecoat of CBC90394 (available from PPG Industries) in three passes with a two-minute flash between coatings. Following the application of the third coat, there is a ten-minute flash dry before applying the clearcoat. The basecoat is not cured before applying the clearcoat.

A clearcoat is then applied over the basecoat using two coats with a two-minute flash dry between coats. Following the second coat, the coated plaque is given a ten-minute flash dry in a dust-free chamber at room temperature. The clear coat is then cured by baking for 20 minutes at 250°F (121°C). The clear coat used in the following examples is PPG's clear coat CUCC-1000, and the dry film thickness is between 0.1 and 0.6 mil.

Finally, a topcoat is applied over the clearcoat. The topcoat utilized in the following examples is PPG Industries EHS 3967. The total topcoat thickness is about 1.8 to 2.0 mil.

Five tests have been utilized to evaluate the adhesivity of the paint to the test plaques.

Test 1: Water Contact Angle

A non-solvent is used to clean the surface of the substrate. For example, for TPO, methyl ethyl ketone is used as the non-solvent. Cleaning can be accomplished by spraying the solvent onto the substrate without wiping which

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simulates actual cleaning conditions. The substrate is allowed to dry in a clean area for about five minutes or until the odor of the non-solvent is no longer present. The clean surface should then be used as soon as possible to avoid contamination. Drops of water and methylene iodide are placed on the clean surface, and for each drop of liquid, the contact angles on both sides of the drop are measured. At least five drops of each liquid are used to obtain at least ten data points for each liquid. The contact angles may be determined on a goniometer or a contact angle analyzer. The average contact angles of the water and methylene iodide are then recorded.

Test 2: Initial Paint Adhesion

The procedure utilized in this test is as specified in ASTM D-3359-87, Method B. The coating on the test plaque is cut lengthwise and crosswise by a razor to form a grid. An adhesive tape (3M's Scotch Brand 898) is pressed onto the grid smoothly but firmly. The tape then is pulled off rapidly at as close to 180° angle as possible. The grid area is then inspected for removal of coating from the substrate or from the previous coating utilizing an illuminated magnifier. For the purposes of this test, a loss of greater than 5% of the coating is considered a failure.

Test 3: Water-Boil Adhesion.

This test is applied as specified in ASTM D-3359-87, Method A with the following variation. The specified cross-hatch is cut into the paint to the substrate. The specimens are placed in a boiling water bath for one hour. At the end of the hour, the test specimens are removed from the bath, cooled to room temperature and air dried. The adhesion of the coating to the substrate is then evaluated as specified in the ASTM method. The test specimen is considered to pass this test if there is a loss of adhesion of 5% or less.

Test 4: Solvent Resistance.

This test also is applied as specified in ASTM D-3359-87, Method A with the following variation. The specified cross-hatch is cut into the paint to the substrate. The specimens are then placed in a solvent mixture of 55% toluene and 45% hexane for one hour. The time to failure is noted. A passing specimen is

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defined as one having been soaked in the solvent for the full 60 minutes with no loss of adhesion. Adhesion is not tested with the tape/pull method in this test.

Test 5: Humidity Chamber.

This test evaluates the adhesion of paint to the polymer blends of the invention under conditions of high humidity using test plaques painted as described above. The painted plaques are placed in the humidity chamber and maintained at 90-100% relative humidity and 100°F (38°C). The plaques are removed from the chamber after 240 hours of exposure and examined. A passing specimen is defined as one that exhibits no marked signs of peeling or adhesion loss (e.g, bubbles, cracking or delamination of paint) from substrate after tape test.

Various features and aspects of the present invention are illustrated further in the examples that follow. While these examples are presented to show one skilled in the art how to operate within the scope of this invention, the examples are not to serve as a limitation on the scope of the invention since such scope is only defined in the claims.

Unless otherwise indicated in the following examples and elsewhere in the specification and claims, all parts and percentages are by weight, temperatures are in degrees Centigrade, and pressures are at or near atmospheric. The physical properties described in the following examples are measured in accordance with ASTM Standard Test Procedures as identified in the following Table I. The sources of the raw materials used in the subsequent examples are identified in Table II.

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TABLE I

<u>Property</u>	<u>ASTM Method</u>
Tensile Strength	D-638
Elongation	D-638
Flexural Strength	D-790
Flexural Modulus (tangent)	D-790
Izod Impact (notched)	D-256*
Izod Impact (unnotched)	D-256
Gardner Impact	D-3209
Heat Deflection Temp.	D-648
Melt Flow Rate	D-1238**

* Method A.

** Condition 230°C/2.16 Kg.

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TABLE II

<u>Company/Grade</u>	<u>Description</u>
Exxon PP 1012	PP Homopolymer
Himont Profax 6523	PP Homopolymer
Shell 5A64	PP Homopolymer
Shell 7C54H	PP Copolymer
Himont Profax 8623	PP Copolymer
Aristech 4040F	PP Copolymer
Eastman Epolene E-43	Maleated PP wax
Chevron SP 2205	Ethylene Methyl Acrylate Copolymer (% MA=20)
Chevron SP 2260	" (% MA-24)
Exxon XS-55-48	" (% MA-28)
Atochem Lotryl 3610	" (% MA-29)
Colonial Rubber EPP 99GA02BK	A masterbatch of a conductive carbon black in polypropylene
Union Carbide DHDA 7704	A masterbatch of a conductive carbon black, in EEA copolymer
Cyprus, Stellar 600	Talc
R.T. Vanderbilt Select-A-Sorb	Talc
Witco, Drapex 4.4	Epoxidized Tall Oil
Ciba-Geigy Irganox 1010	Antioxidant
Exxon Vistalon 719	EP Rubber (pellets)
Exxon Vistalon 3708	EPDM Rubber (pellets)
Shell Kraton G1652	SEBS Block Copolymers
Dutral PM0GPLE	EPR Masterbatch in HDPE from Enimont
DuPont Nacrel 980	Ethylene methacrylic acid copolymer
BP Polybond 1101	Acrylic acid grafted polypropylene

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TABLE II (Cont'd)

<u>Company/Grade</u>	<u>Description</u>
Quantum Vynathene 90230	EVA Copolymer (ethylene vinyl acetate copolymer)
Shell Kraton FG 1901X	Maleated SEBS block copolymer
Ferro 72-886-1	Gray liquid color concentrate
Ferro CH 21119-800	Maroon liquid color concentrate

Examples 1-2

Paintable TPO compositions, which consisted of PP homo- and copolymers, ethylene methyl acrylate copolymers, maleated PP wax, carbon black masterbatch, epoxidized tall oil as plasticizer, talc filler and an antioxidant, were prepared in a laboratory Banbury mixer (300-325 °F, 150 rpm, 6-8 minutes total mixing time, 1100 gram as a batch size) as described in Table III. After injection molding, the test specimens were tested for physical properties and paintability. The results also are reported in Table III.

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TABLE III

<u>Composition (wt. %)</u>	<u>Ex. 1</u>	<u>Ex. 2</u>
Exxon PP1012	9.9	9.9
Shell 7C54H	22.0	22.0
Epolene E-43	9.0	9.0
Chevron SP2205	32.0	32.0
	12.0	12.0
Witco Drapex 4.4 5.0	5.0	
Select-A-Sorb	10.0	10.0
Irganox 1010	0.1	0.1
<u>Physical Properties:</u>		
Melt Flow (g/10 min)	21.6	21.6
Flexural Modulus		
(10 ⁵ psi)	1.00	1.00
Gardner Impact, 25°C		
(in. lb.)	104	104
Linear Shrinkage		
(in/in %)	0.47	0.47
<u>Paintability Evaluation:</u>		
Use of Adhesion Pro-		
moter	Yes	No
Water Contact		
Angle (°)	84.0±1.4	84.0±1.4
Initial Adhesion	pass	pass
Water Boil Test	fail	pass
Solvent Resistance	---	pass
Humidity Test	---	pass

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The results of the paintability evaluation showed that improved adhesion was achieved for the painted plaques that did not contain the adhesion promoter when compared to the painted counterparts that contained an additional coat of adhesion promoter.

Examples 3-6

Blends of polypropylene homopolymer, polypropylene copolymer, maleated polypropylene wax and various ethylene-methylacrylate copolymers are prepared in accordance with the formulations identified in Table IV. The blends of Examples 3, 5 and 6 are prepared in a Banbury mixer and the blend of Example 4 is prepared in a single screw extruder at a temperature of about 190-250°C, and injection-molded into test specimens. The physical properties of the test specimens are summarized in Table IV.

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TABLE IV

<u>Composition</u>		<u>Ex. 3</u> <u>(%Wt)</u>	<u>Ex. 4</u>	<u>Ex. 5</u> <u>(%Wt)</u>	<u>Ex. 6</u> <u>(%Wt)</u>
PP Homopolymer (PP1012)	9.9	9.9	9.9	9.9	
PP Copolymer (7C54H)		22.0	22.0	22.0	22.0
Maleated PP Wax (Epolene E-43)		9.0	9.0	9.0	9.0
Carbon Black*		12.0	12.0	12.0	12.0
Epoxidized Tall Oil (Drapex 4.4)		5.0	5.0	5.0	5.0
Talc (Select-A-Sorb)		10.0	10.0	10.0	10.0
Antioxidant (Irganox 1010)	0.1	0.1	0.1	0.1	
E-MA Copolymer (SP2205)		32.0	---	---	---
E-MA Copolymer (SP2260)		---	32.0	---	---
E-MA Copolymer (XS-55.48)		---	---	32.0	---
E-MA Copolymer (Lotryl 3610)		---	---	---	32.0

Physical Properties

Melt Index, 230°C/2160 g (g/10 min)	21.6	11.2	31.2	23.0
Flexural Modulus (10 ⁵ psi)	1.00	0.82	0.66	0.69
Izod Impact, notched 1/8" (ft.lb./in.)	---	---	7.20	8.20
Gardner Impact 23°C (in.lb)	104	94	187	167
Linear Mold Shrinkage (in./in./%)	0.47	0.37	0.24	0.31

* Masterbatch of a conductive carbon black, from Colonial Rubber Works.

Test specimens from Examples 3-6 were also painted in accordance with the procedure described above and the painted specimens were evaluated utilizing the paintability evaluation tests described above. The results of the paintability evaluation are summarized in the following Table V.

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TABLE V

Paintability Evaluation*

<u>Test</u>	<u>Results</u>			
<u>Product of:</u>	<u>Ex. 3</u>	<u>Ex. 4</u>	<u>Ex. 5</u>	<u>Ex. 6</u>
Water Contact Angle (°)	84.0±1.4	---	78.6±0.9	81.6±0.9
Initial Adhesion	Pass	Pass	Pass	Pass
Water Boil Test	Pass	Pass	Pass	Pass
Solvent Resistance	Pass	Pass	Pass	Pass
Humidity Test	Pass	---	Pass	Pass

* Contains no adhesion promoter.

As can be seen from the above results, the blends of Examples 3-6 exhibited similar and excellent paintability results with the different ethylene-methacrylate copolymers obtained from different suppliers.

Examples 7-9

These examples are similar to Example 3 except that the composition of Example 8 contains 6% by weight of a carbon black masterbatch which is different from that used in Example 3, namely, conductive carbon black XC-72 in EEA copolymer (Union Carbide grade No. DHDA 7704) and the composition of Example 9 contains no carbon black, but additional polypropylene copolymer.

The components identified for the compositions of Examples 7-9 are mixed in a Banbury mixer and injection molded to form test plaques which are thereafter cleaned and painted in accordance with the procedure described above. The components and amounts utilized in the preparation of the compositions of Examples 7-9 are identified in Table VI. The physical properties of the blended polymer compositions and extruded articles also are summarized in Table VI. The results of the paintability evaluation on the molded test pieces of Examples 7-9 are reported in Table VII. As can be seen from the results reported in Table VII, the

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blended polymer compositions of Examples 7-9 exhibit excellent paintability with or without carbon black masterbatch.

TABLE VI
Blended Polymer Compositions

<u>Composition</u>		<u>Ex. 7</u> <u>(%Wt)</u>	<u>Ex. 8</u> <u>(%Wt)</u>	<u>Ex. 9</u> <u>(%Wt)</u>
PP Homopolymer (PP1012)	9.9	9.9	9.9	
PP Copolymer (Profax 8623)		22.0	28.0	34.0
E-MA Copolymer (SP2205)		32.0	32.0	32.0
Maleated PP Wax (Epolene E-43)		9.0	9.0	9.0
Carbon Black ¹		12	---	---
Carbon Black ²		---	6.0	---
Epoxidized Tall Oil (Drapex 4.4)		5.0	5.0	5.0
Talc (Select-A-Sorb)		10.0	10.0	10.0
Antioxidant (Irganox 1010)	0.1	0.1	0.1	

Physical Properties

Melt Index, 230°C/2160 g (g/10 min)	18.2	22.0	26.2
Flexural Modulus (10 ⁵ psi)	0.83	0.79	0.84
Izod Impact, notched 1/8" (ft.lb./in.)	2.60	6.90	2.30
Gardner Impact 23°C (in.lb)	133	153	149
Gardner Impact -20°C (in.lb.)	---	27	---
Linear Mold Shrinkage (in./in./%)	0.50	0.39	0.53

1 Masterbatch from Colonial Rubber Works.

2 Masterbatch in EEA copolymer (Union Carbide DHDA 7704).

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TABLE VII

Paintability Evaluation*

<u>Test</u>	<u>Ex. 7</u>	<u>Results</u>	
		<u>Ex. 8</u>	<u>Ex. 9</u>
Water Contact Angle	78.0±2.7	81.8±0.8	80.4±0.6
Initial Adhesion	Pass	Pass	Pass
Water Boil Test	Pass	Pass	Pass
Solvent Resistance	Pass	Pass	Pass
Humidity Test	Pass	Pass	Pass

* No adhesion promoter used.

Example 10

This example is similar to Example 7 except that Stellar 600 talc is used in place of the Select-A-Sorb. A blend is prepared in a Banbury mixer. The amounts of components for this example is identified in the following Table VIII. The blended components are injection-molded to form test plaques which are thereafter cleaned and painted in accordance with the procedure described above. The physical properties of the blended polymer composition and extruded articles are also summarized in Table VIII. The results of the paintability evaluation on the molded test pieces of Example 10 are reported in Table IX.

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TABLE VIII

<u>Composition</u>	<u>Ex. 10</u> <u>(%Wt)</u>
PP Homopolymer (PP1012)	9.9
PP Copolymer (Profax 8623)	22.0
Maleated PP Wax (Epolene E-43)	9.0
Carbon Black Masterbatch (Colonial)	12.0
Epoxidized Tall Oil (Drapex 4.4)	5.0
Antioxidant (Irganox 1010)	0.1
E-MA Copolymer (SP2205)	32
Stellar 600 Talc	10
<u>Physical Properties</u>	
Melt Index, 230°C/2160 g (g/10 min)	21.2
Flexural Modulus (10 ⁵ psi)	0.86
Izod Impact, notched 1/8" (ft.lb./in.)	3.30
Gardner Impact 23°C (in.lb)	125
Linear Mold Shrinkage (in./in./%)	0.50

TABLE IX

Paintability Evaluation*

<u>Test</u>	<u>Results</u>
<u>Product of:</u>	<u>Ex. 10</u>
Water Contact Angle (°)	80.2±1.1
Initial Adhesion	Pass
Water Boil Test	Pass
Solvent Resistance	Pass
Humidity Test	Pass

* No adhesion promoter used.

Example 11-13

These examples utilize components similar to those used in Example 3 except that the amounts of some of the components are different and the composi-

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tions contain modifiers (IV) as described above. The components and amount of components utilized in these examples are summarized in the following Table X. The components are mixed in a Banbury mixer and injection molded to form test plaques where are thereafter cleaned and painted in accordance with the procedure described above. The physical properties of the blended polymer compositions and extruded articles also are summarized in Table X. The results of the paintability evaluation on the molded test pieces are reported in Table XI. As can be seen from the results reported in Table X, the blended polymer compositions containing a modifier as defined in the present specification and claims exhibit improved low temperature impact properties.

TABLE X

3*		Ex. 11		Ex. 12		Ex. 13Ex.	
		<u>(%Wt)</u>		<u>(%Wt)</u>		<u>(%Wt)</u> <u>(%Wt)</u>	
<u>Composition</u>	PP Homopolymer (PP1012)	5.3	5.3	5.3	9.9		
	PP Copolymer (7C54H)		11.6	11.6	11.6	22.0	
	Maleated PP Wax (Epolene E-43)		9.0	9.0	9.0	9.0	
	Carbon Black Masterbatch (Colonial)		12.0	12.0	12.0	12.0	
	Epoxidized Tall Oil (Drapex 4.4)		5.0	5.0	5.0	5.0	
	Talc (Select-A-Sorb)		10.0	10.0	10.0	10.0	
	Antioxidant (Irganox 1010)	0.1	0.1	0.1	0.1		
	E-MA Copolymer (SP2205)		32.0	32.0	32.0	32.0	
	EPR-(Vistalon 719)		15.0	----	----	----	
	EPDM (Vistalon 3708)		----	15.0	----	----	
	SEBS Block Copolymer (Kraton G1652)		----	----	15.0	----	
<u>Physical Properties</u>							
	Melt Index, 230°C/2160 g (g/10 min)		12.2	12.4	17.4	21.6	
	Flexural Modulus (10 ⁵ psi)		0.58	0.55	0.57	1.00	
	Gardner Impact 23°C (in.lb.)		126	132	180	104	
	Gardner Impact -20°C (in.lb.)		41	95	136	6	
	Linear Mold Shrinkage (in./in./%)		0.30	0.27	0.25	0.47	

* Ex. 1 results shown for comparison.

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TABLE XI**Paintability Evaluation***
Results

<u>Test</u>				
<u>Product of:</u>	<u>Ex.11</u>	<u>Ex.12</u>	<u>Ex.13</u>	<u>Ex.3</u>
Water Contact Angle (°)	84.4±0.6	78.6±3.1	85.4±1.5	84±1.4
Initial Adhesion	Pass	Pass	Pass	Pass
Water Boil Test	Pass	Pass	Pass	Pass
Solvent Resistance	Pass	Pass	Pass	Pass
Humidity Test	Pass	Pass	Pass	Pass

* No adhesion promoter used.

Examples 14-16

These examples are prepared in the same manner as Example 7 except that other polymers are utilized in place of the maleated polypropylene (E-43). The components and amounts of components are identified in the following Table XII, and the physical properties of the blended polymer compositions and extruded articles are also summarized in Table XII. The results of the paintability evaluation on the molded test pieces of Examples 14-16 are reported in Table XIII.

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TABLE XII

<u>Composition</u>	<u>Ex.14</u> <u>(%Wt)</u>	<u>Ex.15</u> <u>(%Wt)</u>	<u>Ex.16</u> <u>(%Wt)</u>
PP Homopolymer (PP1012)	9.9	9.9	9.9
PP Copolymer (Profax 8623)	28.0	28.0	28.0
Maleated SP Wax (Epolene E-43)	9.0	---	---
Carbon Black Conc. (DHDA 7704)			
Union Carbide	6.0	6.0	6.0
Epoxidized Tall Oil (Drapex 4.4)	5.0	5.0	5.0
Talc (Select-A-Sorb)	10.0	10.0	10.0
Antioxidant (Irganox 1010)	0.1	0.1	0.1
EMAC (SP2205)	32.0	32.0	32.0
Acrylic acid grafted PP (Polybond 1101)	---	9.0	---
Ethylene-methacrylic acid copolymer (Nucrel 960)	---	---	9.0

Physical Properties

Melt Index, 230°C/2160 g (g/10 min)	22.0	4.7	3.1
Flexural Modulus (10 ⁵ psi)	0.79	0.70	0.43
Gardner Impact 23/°C (in.lb.)	153	193	203
Linear Mold Shrinkage (in./in./%)	0.39	0.46	0.71

TABLE XIIIPatentability Evaluation*

<u>Test</u>	<u>Ex.14</u>	<u>Ex.15</u>	<u>Ex.16</u>
Water Contact Angle	81.8±0.8	91.6±1.1	90.2±0.8
Initial Adhesion	Pass	Fail	Fail
Water Boil Test	Pass	Fail	Fail
Solvent Resistance	Pass	Fail	Fail
Humidity Test	Pass	Fail	Fail

* No adhesion promoter used.

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Examples 17-19

In these examples, the following modifiers are used and compared: EMA copolymer; EVA copolymer, and SEBS-MA graft copolymer. The general procedure for Example 1 is repeated in preparing blended components which are injection-molded to form test plaques. The amounts of the components used in preparing the blended compositions are summarized in Table XIV. The results of the paintability evaluation on the molded test pieces are reported in Table XV.

TABLE XIV

<u>Composition</u>	<u>Ex.17</u> <u>(%Wt)</u>	<u>Ex.18</u> <u>(%Wt)</u>	<u>Ex.19</u> <u>(%Wt)</u>
PP Homopolymer (PP1012)	9.9	9.9	5.24
PP Copolymer (7C54H)	22.0	22.0	11.66
Maleated PP Wax (Epolene E-43)	9.0	9.0	---
Carbon Black	12.0	12.0	12.0
Epoxidized Tall Oil (Drapex 4.4)	5.0	5.0	5.0
Talc (Select-A-Sorb)	10.0	10.0	10.0
Antioxidant (Irganox 1010)	0.1	0.1	0.1
E-MA Copolymer (SP2105)	32.0	---	---
EVA Copolymer (Vynathene 90230)	---	32	32
SEBS-MA (Kraton GF1901X)	---	---	24

TABLE XIIIPaintability Evaluation*

<u>Test</u>	<u>Ex.17</u>	<u>Ex.18</u>	<u>Ex.19</u>
Water Contact Angle			
Initial Adhesion	Pass	Pass	Pass
Water Boil Test	Pass	Pass	Pass
Solvent Resistance	Pass	Pass	Pass

* No adhesion promoter used.

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Examples 20-22

These examples illustrate the preparation of colored polymer compositions which do not have to be painted. In these examples, the injection-molded colored plaques are coated with a clear finish coat in the absence of an adhesion promoter. The general procedure of Example 3 is repeated in preparing these examples. Components and amount of components utilized for preparing the blended polymer compositions are summarized in Table XVI. The compositions are prepared from similar components except for the pigment. Example 20 includes carbon black; Example 21 includes a gray pigment and Example 22 includes a maroon pigment. The results of the evaluation on molded test pieces of these examples coated with a clear acrylic finish are reported in Table XVII.

TABLE XVI

<u>Composition</u>	<u>Ex.20</u> <u>(%Wt)</u>	<u>Ex.21</u>	<u>Ex.22</u> <u>(%Wt)</u>	<u>Ex.22</u> <u>(%Wt)</u>
PP Homopolymer (PP1012)	5.3	8.7	7.7	
PP Copolymer (7C54H)	11.6	19.2		17.2
Maleated PP Wax (Epolene E-43)	9.0	9.0		9.0
Carbon Black	12.0	---	---	
Epoxidized Tall Oil (Drapex 4.4)	5.0	5.0		5.0
Talc (Select-A-Sorb)	10.0	10.0		10.0
Antioxidant (Irganox 1010)	0.1	0.1	0.1	
E-MA Copolymer (SP2205)	32.0	32.0		32.0
SEBS Block Copolymer (Kraton G1652)	15.0	15.0		15.0
Gray Pigment (Ferro 72-886-1)	---	1.0		---
Maroon Pigment (Ferro CH21119-800)	---	---		4.0

TABLE XVII

<u>Test</u>	<u>Ex.20</u>	<u>Ex.21</u>	<u>Ex.22</u>
Water Contact Angle			
Initial Adhesion	Pass	Pass	Pass
Water Boil Test	Pass	Pass	Pass
Solvent Resistance	Pass	Pass	Pass
Humidity Test	Pass	Pass	Pass

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Examples 23-27

These examples demonstrate the improved results obtained with the blended polymer compositions of the invention, and the criticality of the presence of both the elastomeric copolymer (II) and the reaction product (III). (Example C-1 does not contain any elastomer composition (II). Example C-2 does not contain a reaction product (III).) The components and amount of components used are identified in Table XVIII. The components are mixed in a single screw extruder at normal processing conditions of polyolefin systems. The blended compositions were injection molded into test specimens and painted in accordance with the general procedure described earlier. The painted specimens were evaluated utilizing the paintability evaluation tests described earlier, and the results are summarized in the following Table XIX.

TABLE XVIIIExamples

<u>Composition (wt.%)</u>	<u>23</u>	<u>24</u>	<u>25</u>	<u>26</u>	<u>27</u>	<u>C-1</u>	<u>C-2</u>
Shell 5A64		10.3	12.47	14.03	15.59	17.16	20.28
Shell 7C54H		14.6	19.43	22.87	26.31	29.74	36.62
Chevron SP2205		32	25	20	15	10	0
Dutral PM06-PLE		23	23	23	23	23	23
Select-A-Sorb (Talc)		10	10	10	10	10	10
Epolene E-43		9	9	9	9	9	0
DHDA 7704		1	1	1	1	1	1
Irganox 1010 Stabilizer		0.1	0.1	0.1	0.1	0.1	0.1

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TABLE XIXPaintability Results*

<u>Test</u>	<u>23</u>	<u>24</u>	<u>25</u>	<u>26</u>	<u>27</u>	<u>C-2</u>	<u>C-1</u>
Initial Adhesion	pass	pass	pass	pass	fail	fail	fail
Water Boil Test	pass	pass	pass	fail	fail	fail	fail
Solvent Resistance	pass	pass	pass	pass	fail	fail	fail

* No adhesion promoter used.

Although Examples 23-27 do not contain a plasticizer (epoxidized tall oil), these were easily processable and directly paintable.

While the invention has been explained in relation to its preferred embodiments, it is to be understood that various modifications thereof will become apparent to those skilled in the art upon reading the specification. Therefore, it is to be understood that the invention disclosed herein is intended to cover such modifications as fall within the scope of the appended claims.

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Claims

1. A blended polymer composition comprising:
 - (I) at least one crystalline alpha-olefin polymer;
 - (II) from about 5 to about 60% by weight of at least one elastomeric copolymer of an alpha-olefin and an alpha,beta-unsaturated carboxylic acid ester;
 - (III) from about 1 to 30% by weight of the product of the reaction of an alpha-olefin polymer and an alpha,beta-olefinically unsaturated carboxylic acid reagent; and
 - (IV) from about 0 to about 40% by weight of at least one impact modifier selected from the group consisting of
 - (a) an ethylene-propylene copolymer elastomer which is different from the alpha-olefin polymer (I);
 - (b) an elastomer which is a copolymer of a vinyl aromatic compound and a conjugated diene; and
 - (c) a selectively hydrogenated block copolymer of a vinyl aromatic compound and a conjugated diene.
2. A blended polymer composition of claim 1 comprising from about 25 to about 50% by weight of the alpha-olefin polymer (I).
3. The polymer composition of claim 1 wherein the alpha-olefin polymer (I) is the polymer of an alpha-olefin containing from 2 to about 6 carbon atoms.
4. The polymer composition of claim 1 wherein the alpha-olefin polymer (I) is polyethylene, polypropylene, or mixtures thereof.
5. The polymer composition of claim 1 wherein (II) is a copolymer of an alpha-olefin and an acrylic acid ester.
6. The polymer composition of claim 5 wherein the ester is a methyl, ethyl or butyl ester.
7. The polymer composition of claim 1 wherein the alpha-olefin polymer reacted in (III) is an ethylene or propylene polymer.

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8. The polymer composition of claim 1 wherein the carboxylic acid reagent reacted in (III) is a dicarboxylic acid reagent.

9. The polymer composition of claim 1 wherein the alpha-olefin polymer (I) is polypropylene and the polymer composition comprises from about 5 to about 20% by weight of a modifier (IV).

10. The polymer composition of claim 1 also containing

(V) from about 0.5 to about 10% by weight of at least one pigment.

11. The blended polymer composition of claim 1 also containing

(VI) at least one filler, fiber, or mixtures thereof.

12. A blended polymer composition comprising:

(I) from about 25 to about 50% by weight of at least one alpha-olefin polymer derived from ethylene, propylene, or mixtures thereof;

(II) from about 20 to about 40% by weight of at least one copolymer of ethylene or propylene and an acrylic ester;

(III) from about 5 to about 15% by weight of the product of the reaction of a polyethylene or polypropylene with an alpha,beta-olefinically unsaturated dicarboxylic acid reagent; and

(IV) from zero to about 20% by weight of at least one modifier selected from the group consisting of

(a) an ethylene-propylene copolymer rubber which is different from the alpha-olefin polymer (I);

(b) a copolymer of a vinyl aromatic compound and a conjugated diene; and

(c) a selectively hydrogenated block copolymer of a vinyl aromatic compound and a conjugated diene.

13. The polymer composition of claim 12 wherein the alpha-olefin polymer (I) is a polypropylene.

14. The polymer composition of claim 12 wherein the copolymer (II) is a copolymer of ethylene and a methyl, ethyl or butyl acrylate.

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15. The polymer composition of claim 12 wherein (III) is the product of the reaction of a polypropylene and a dicarboxylic acid reagent selected from maleic acid and maleic anhydride.

16. The polymer composition of claim 12 containing from about 5 to about 25% by weight of at least one modifier (IV).

17. The polymer composition of claim 12 also containing
(V) from about 0.5 to about 10% by weight of at least one pigment.

18. The polymer composition of claim 17 wherein the pigment is a carbon black.

19. The polymer composition of claim 12 also containing
(VI) at least one filler, fiber or mixtures thereof.

20. A blended polymer composition comprising:

(I) from about 20 to about 60% by weight of at least one polymer of ethylene or propylene;

(II) from about 20 to about 40% by weight of at least one copolymer of ethylene or propylene with a methyl, ethyl or butyl acrylate;

(III) from about 5 to about 15% by weight of the product of the reaction of ethylene or propylene with maleic acid or maleic anhydride;

(IV) from about zero to about 20% by weight of at least one modifier selected from the group consisting of

(a) an ethylene-propylene copolymer rubber which is different from the alpha-olefin polymer (I);

(b) a copolymer of a vinyl aromatic compound and a conjugated diene; and

(c) a selectively hydrogenated block copolymer of a vinyl aromatic compound and a conjugated diene; and

(V) from about 0.5 to about 5% by weight of carbon black, and

(VI) from about 0 to 5% by weight of a plasticizer.

INTERNATIONAL SEARCH REPORT

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A. CLASSIFICATION OF SUBJECT MATTER

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US CL :525/78

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B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 525/78

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

N.A.S. PLATZER, ED, "Copolymers, Polyblends and Composites, published 1975, by American Chemical Society (Washington, DC).

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US,A, 4,957,968 (ADUR ET AL.) 18 September 1990, See the entire document.	1-20
Y	N.A.J. PLATZER, EDITOR, "Copolymers, Polyblends, and Composites", 1975, by American Chemical Society (Washington, DC), See pages 186 and 191.	1-20



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents:	*T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
A document defining the general state of the art which is not considered to be part of particular relevance	*X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
E earlier document published on or after the international filing date	*Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
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O document referring to an oral disclosure, use, exhibition or other means	
P document published prior to the international filing date but later than the priority date claimed	

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